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Determining the Ages of Recent Sediments Using Measurements of Trace Radioactivity

Abstract

Laboratory analyses of sediment cores can determine sedimentation rates and the calendar dates associated with various depths within sediments. These chronology results can be used for characterising the deposition environment of a water system, which is pertinent to the planning of dredging operations. The methods are particularly useful for water systems which contain buried toxic substances. Measurements of different radioactive species give chronology information for time frames from a half year to 100 years before the present. Recent studies in the 1990s where this method has been applied include the Kalamazoo River in Michigan, the Housatonic River in Connecticut, the Passaic River in New Jersey, the Hudson River and Grasse River in New York. This paper presents the practical application of three geochronology methods which have been used.

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Introduction

The accumulation of sediment in a water system is one of the most common situations which requires dredging. Consequently, a knowledge of sedimentation rates is often useful in planning dredging operations. The pattern of sedimentation rates can be used to calculate when various areas will require work. This information can also be used to plan the time intervals between successive operations. In shipping channels and other locations which are frequently dredged, sedimentation rates can be calculated from the history of periodic depth surveys. There are instances, however, for which no history of depth surveys exist.

In recent years there has been a growing emphasis on characterisation and remediation of contaminated waterways. Often a long stretch of river and the associated terminal estuary has been contaminated by industries over a period of decades. The toxic substances released by these industries become progressively buried in the sediments, such that profiles of these substances in the sediment become a record of the contamination process. These water systems may have only a fragmentary history of depth surveys, or none at all. Such water systems require another method to measure sedimentation rates and to determine the calendar dates associated with buried toxic substances.

Chronology information of this kind is useful in determining which industries caused the contamination, for industrial production or release records can be compared to the dates of buried materials. Chronology information is also useful for determining whether buried substances are migrating or degenerating.
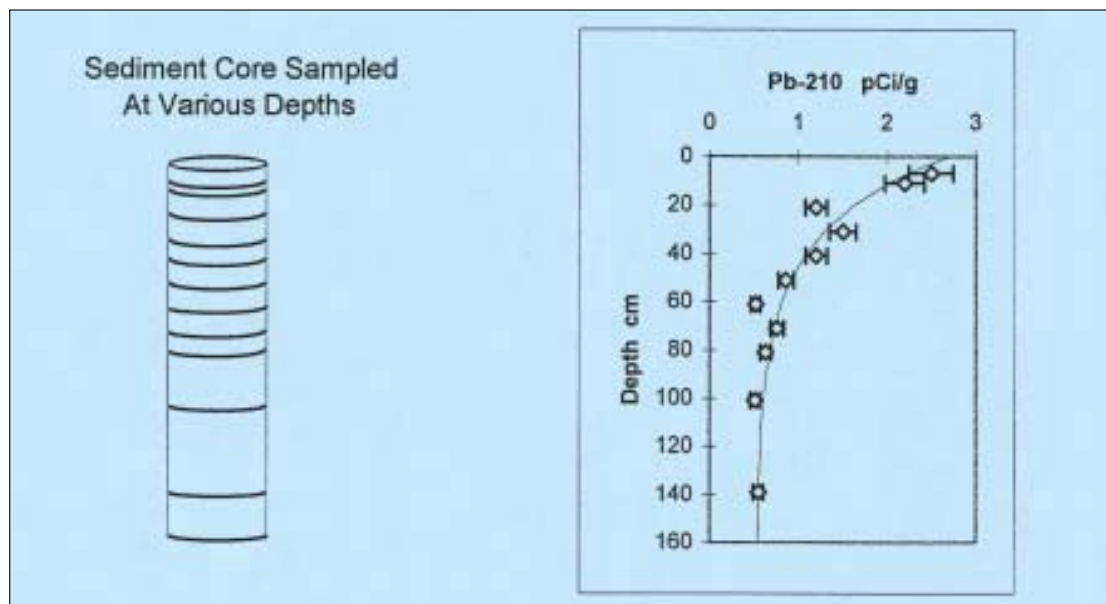
Figure 1. A profile of Pb-210 concentrations measured in a sediment core. Results are expressed in picocuries per gram (pCi/g). The picocurie is a small unit of radioactivity equivalent to 2.22 nuclear transitions per minute.

Trace quantities of radioactivity, primarily of natural origin, are found in most substances. The earth, the oceans, the atmosphere and living things have always contained a number of radioactive species. Scientific methods for measuring recent sedimentation rates by analysing trace quantities of radioactivity were developed by universities beginning more than thirty years ago (Ref. 1). These methods were subsequently expanded and applied to lakes, ocean environments, and rivers (Ref. 2-5).

With increasing interest in the environment, the techniques were applied to contaminated river and estuary systems (Ref. 6-8). The procedures involve taking sediment cores and analysing samples at various depths. The distribution of natural or artificial radioactive species in the cores can often be interpreted to produce a chronological history of the sediments and their associated contaminants.

Present studies of contaminated river systems often include the taking of a pattern of sediment cores to characterise the site. The number of cores may be fewer than 10 or greater than 100. Cores are often 4 to 10 cm in diameter and 1 to 7 m in length. The cores are generally cut lengthwise to enable a sedimentary geologist to make a visual study of the sediments, sometimes including grain size measurements. Transverse sections of the cores are then sampled and sent to a laboratory to measure the concentrations of contaminants, resulting in profiles of the contaminants as a function of depth. Other sections of the core, often 2 cm in thickness, are sent to a radiochemistry laboratory to measure trace radioactivity at various depths. The radioactivity data are interpreted to determine sedimentation rates and the dates associated with different

Table I. Pb-210 concentrations plotted in Figure 1.

Depth cm	Pb-210 pCi/g
6 – 8	2.5 +– 0.2
10 – 12	2.2 +– 0.2
20 – 22	1.2 +– 0.1
30 – 32	1.5 +– 0.1
40 – 42	1.2 +– 0.2
50 – 52	0.86 +– 0.12
60 – 62	0.52 +– 0.08
70 – 72	0.76 +– 0.10
80 – 82	0.63 +– 0.13
100 – 102	0.51 +– 0.11
138 – 140	0.55 +– 0.12

Tolerances of the measurements are based on detection uncertainties at the 2 sigma (95% confidence) level.

depths in the sediment. The chronology data are then linked to the contaminant profiles in order to characterise the site.

The Teledyne Environmental laboratory has supported many site characterisations since 1977 by performing radiochemical analyses of core samples and interpreting the sediment chronology. Work of this kind has been performed for 23 engineering firms and for 15 universities. Recent studies in the 1990s include the Kalamazoo River in Michigan, the Housatonic River in

Figure 2. Radiochemcial separations are performed to isolate bismuth or polonium from sediment samples. The purified elements are then analysed in radioactivity detectors.

Connecticut, the Passaic River in New Jersey, the Hudson River and Grasse River in New York. This paper presents the practical application of three geochronology methods which have been used.

CHRONOLOGY STUDIES ON THE 100 YEAR TIME FRAME: THE PB-210 METHOD

The Pb-210 (lead-210) method performs best in relatively quiet deposition areas such as marsh lands (Ref. 9), bays, lakes (Ref. 4) and the backwaters of river systems. For example, in the Passaic River (New Jersey), more than 100 sediment cores were analysed for Pb-210. Of those, the three cores taken from quiescent tributaries exhibited more regular profiles. Fast flowing rivers may produce intermittent deposition which is better measured by the Cs-137 (cesium-137) method described in the next section. Nevertheless, the Pb-210 method is often used in conjunction with the Cs-137 method in active rivers in order to obtain maximum chronology information.

Lead-210 is a natural radioactive form of lead which is found in small quantities in most soils as part of the uranium decay series. It is also produced as natural fallout from the atmosphere by radioactive decay of Rn-222 (radon-222) gas. Minute quantities of Pb-210 fall constantly onto the earth's surface. This material accompanies and mixes with sediments which accumulate at the bottoms of water systems. For a given locality, the supply of Pb-210 is often at a steady rate,

Figure 3. Purified bismuth derived from a sediment sample is loaded into a low level beta-particle detector. The measured radioactivity of Bi-210 is used to calculate the concentration of Pb-210.

being derived from direct deposition, from upstream transport, and from decay of Rn-222 in the water. The result is a relatively high concentration of Pb-210 in the shallow sediments.

Figure 1 and Table I show an example of Pb-210 concentrations found at various depths in a sediment core. The measurements were made by cutting 2 cm thick sections from a core, drying the samples in a laboratory oven, then performing chemical separations to isolate the radioactive elements present (Figure 2). The purified elements are placed in sensitive radioactivity detectors to measure their radioactive concentrations
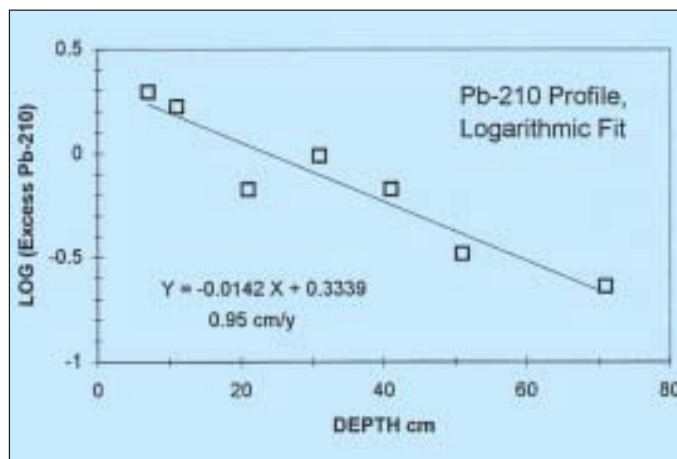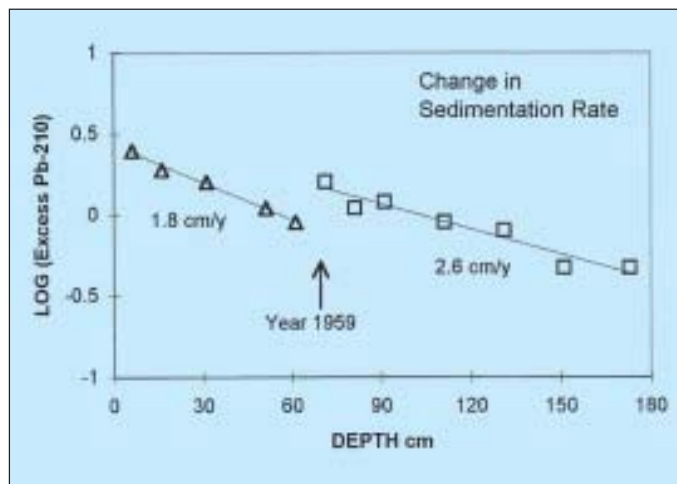
Figure 4. A logarithmic fit to the data presented in Table I and Figure 1. The "excess" Pb-210 concentration is caused by natural fallout. It is calculated in this case by subtracting the average of the two deepest measurements (0.53 pCi/g).

Figure 5. Two distinct sedimentation rates were found in a tributary core sample when an ox bow (loop) was cut off.

(Figure 3). Lead-210 is often measured indirectly by analysing its radioactive decay products Bi-210 (bismuth-210) or Po-210 (polonium-210).

The Pb-210 profile in Figure 1 shows relatively high concentrations in the surface sediment caused by natural fallout. There is decreasing trend with depth, finally achieving a constant level which is inherent in the sediment itself. The decreasing trend is caused by radioactive decay of fallout Pb-210 with time. Deeper levels in a core correspond to earlier times, so that radioactive decay is manifested as decreasing concentration with depth. This is the basis for determining sedimentation rates by the Pb-210 method.

In Figure 4, the logarithm of the "excess Pb-210" derived from natural fallout (the measured concentration minus the constant concentration at deeper levels) is plotted against depth. The linear trend of this plot shows that excess Pb-210 concentration varies logarithmically with depth. This situation occurs because radioactive decay is an exponential process: under simple decay conditions, the logarithm of a radioactive concentration decreases linearly with time. The decay of Pb-210 translates into a logarithmic decrease in excess Pb-210 concentration with depth in the simplest case where the sedimentation rate and the rate of Pb-210 supply are steady, and the upper sediments are nearly uniform in physical properties and intrinsic uranium-series content. Although more complex cases have been studied, simple logarithmic profiles (or segments of profiles) are often found in sediments and can be interpreted usefully.

The equation of the fitted line is shown in Figure 4. The slope of this line can be used to calculate the sedimentation rate, knowing the radioactive decay coefficient of Pb-210. When base 10 logarithms are used and the depths are expressed in cm, the sedimentation rate in cm per year equals -0.01352 / slope. In the case shown, the ratio -0.01352 / -0.0142 leads to a calculated sedimentation rate of 0.95 cm/y.

Because of radioactive decay, excess Pb-210 (derived from natural fallout) is generally detectable to 100 years before the present. At depths corresponding to 100 years or older, the excess Pb-210 has decayed away and the measured concentration represents the background level which is characteristic of the sediment itself. If a logarithmic curve is fitted to a complete Pb-210 profile from the surface to the 100 year level, therefore, the sedimentation rate derived from the slope of the line represents an average over a 100 year time frame. Sometimes shorter segments of profiles are analysed, as will be shown, which lead to sedimentation rates averaged over shorter time periods.

Once a steady sedimentation rate has been derived from a Pb-210 profile, calendar dates at various levels in the sediment are easily calculated. For a given depth, the time interval between the deposition date and the core sampling date is equal to the depth divided by the sedimentation rate. This interval is subtracted from the year in which the core was taken. In the present example, if the core sample were taken in 1998, the date associated with 50 cm depth would be:
1998 - 50/0.95 = 1945.

The "steady" sedimentation rate illustrated does not imply that the rate is strictly constant. The 2 cm increments taken from this core encompass a time period of 2.1 years (2 cm increment / 0.95 cm/y = 2.1 y). Therefore, seasonal or short-term fluctuations of the sedimentation rate would be averaged by the sampling. This shows that logarithmic trends of Pb-210 can be produced even with short-term variations of the sedi-

mentation rate. Similarly, the rate of Pb-210 supply to the sediment does not need to be strictly constant, but steady when averaged over periods of about 2 years in this example.

An unusual case of Pb-210 chronology is shown in Figure 5. This sediment core is derived from a meandering tributary leading into one of the Great Lakes, which formed an ox bow (loop) which was subsequently cut off, straightening its path. Two logarithmic lines with different slopes are shown which indicate two different sedimentation rates. The date when the stream changed course is calculated from the depth of the slope break and from the shallower sedimentation rate. The discontinuity in profiles occurs near 66 cm depth. Because the core was taken in 1996, the date of this event is calculated as 1996 - 66/1.8 = 1959. Cases have been reported where multiple straight line segments are found on logarithmic plots of Pb-210 concentration versus depth.

Figure 6 illustrates the variation in performance of the Pb-210 method. Both logarithmic Pb-210 profiles were obtained in the same river system (the Passaic River) and both indicate nearly the same sedimentation rate. The upper profile exhibits significantly more scatter of the data points which is indicated by its lower value of the correlation coefficient, R2. This scatter may be caused by intermittent deposition during storm, flood and seasonal events. Scatter of Pb-210 data are often associated with layers of sand found between layers of silt. The sand layers, produced by high energy erosion events, often have low values of Pb-210. Profiles which exhibit significant scatter of the data imply greater uncertainty of the sedimentation rates, such as the example of the Passaic River given above. For this reason, the Pb-210 method is often augmented or replaced by the Cs-137 method for fast flowing rivers, as described in the next section.

The Pb-210 method is usually applied to sedimentation rates greater than 0.1 cm/y, which are of primary interest in dredging studies. In the case of a 0.1 cm/y sedimentation rate, the 100 year time frame would produce excess Pb-210 over the shallowest 10 cm of sediment. Often the first few cm of sediments produce anomalous Pb-210 trends, however, which are difficult to interpret. These anomalous measurements may be caused by mixing of the sediments by biological organisms or by physical processes. In addition, the density of sediments may vary considerably in this region, making the application of the simplest model invalid. Often Pb-210 data in the first few centimetres depart from the logarithmic trend found at greater depths, and these data points are excluded in the fitting of logarithmic profiles.

For high sedimentation areas such as bends in river channels and in river deltas, sedimentation rates of 10
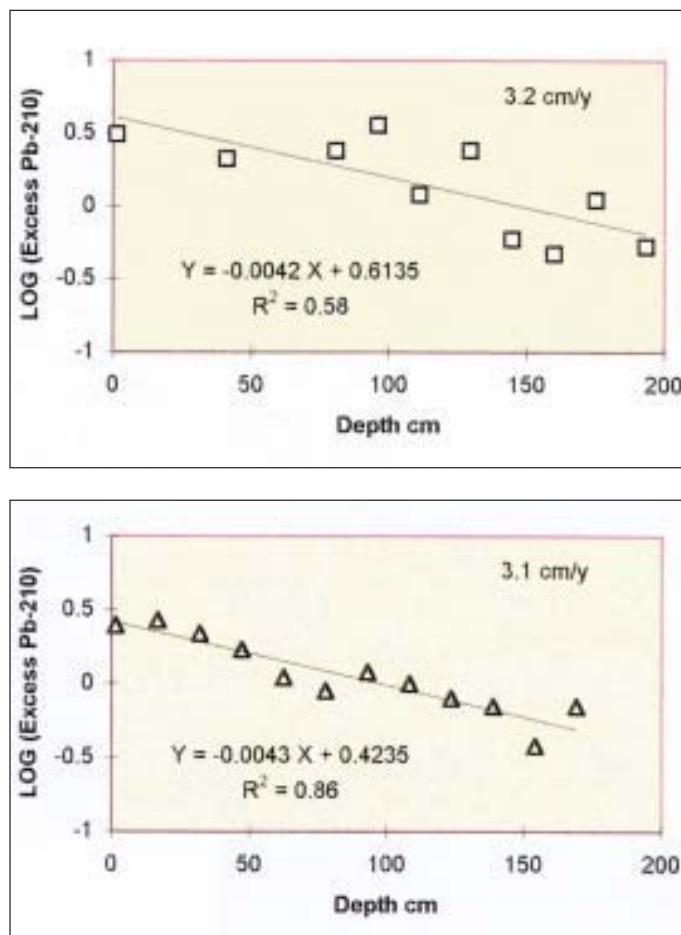

Figure 6. Logarithmic Pb-210 profiles from cores taken at two locations in the same river system (Passaic River). The calculated sedimentation rates are nearly equal but the lower plot exhibits greater certainty because the profile shows less scatter of the data points.

cm/y or greater may be found. At 10 cm/y, excess Pb-210 would be produced to a depth of 10 m if no dredging were performed. Cores of this length are rarely taken in surveys of rivers or estuaries, and sedimentation rates are calculated based on shorter, incomplete cores.

CHRONOLOGY STUDIES OVER THE LAST 45 YEARS: THE CS-137 METHOD

Another powerful chronology method is based on the fallout of Cs-137 (cesium-137) from the atmospheric testing of nuclear weapons. This fission product has been in the atmosphere since the early days of the nuclear age and continues to deposit on the earth in small quantities today. Profiles of Cs-137 in sediments indicate its deposition history which can be interpreted to assign calendar dates. Measurements can be
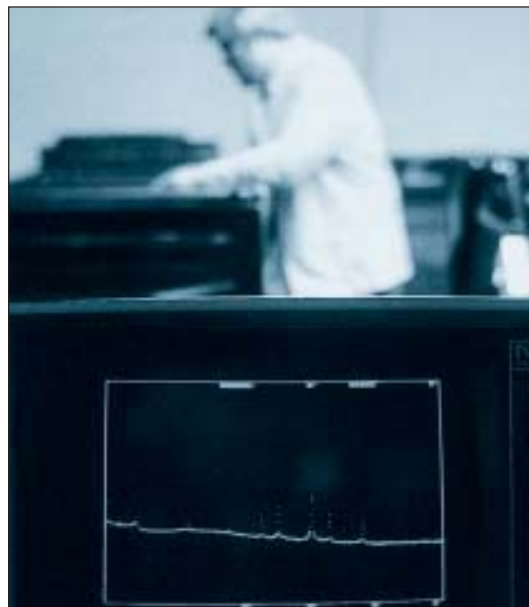
performed in the laboratory by direct analyses of the gamma radiation from a sediment sample (Figures 7 and 8) without any chemical processing.

The Cs-137 method is fundamentally different from the Pb-210 method in that it provides date "markers" rather than concentration slopes which can be interpreted. The first appearance of Cs-137 in sediments generally marks the year 1954, for that is the year when

concentrations generally achieved detectable levels. Thus, if Cs-137 is detected at a given depth, the date is interpreted to be 1954 or afterward. The level in the sediment at which Cs-137 is first detected is called the Cs-137 "horizon", following geological terminology.

If a series of analyses are made at various depths in a sediment core, another Cs-137 marker is often found in the form of a concentration maximum at the year 1963. This is caused by the increase in nuclear testing in the late 1950s and early 1960s, followed by a subsequent decrease in testing.

Figure 9 and Table II show a Cs-137 profile measured in a sediment core in 1995. The profile shows a Cs-137 horizon near 48 cm depth. This marker can be used to calculate a sedimentation rate as follows: (48 cm depth) / (41 y between 1954 and 1995) = 1.2 cm/y. The profile also shows a Cs-137 maximum near 37 cm depth. This marker can also can be used to calculate a sedimentation rate: (37 cm depth) / (32 y between 1963 and 1995) = 1.2 cm/y. In this case, the same sedimentation rate is calculated from both markers.

This is not always the case, however, because events such as unusual floods could occur between the years 1954 and 1963, causing the two calculations to differ. A study in the Delaware River estuary showed sedimentation rates between 1954 and 1963 to be twice as high as sedimentation rates after 1963 because of storm events in the earlier interval (Ref. 9).

Figure 7. A sample prepared in a standard cylindrical container is placed on a germanium-lithium diode detector to measure its gamma radiation.

Figure 8. The gamma ray energy spectrum shown on this screen represents several radioactive species being analysed by the germanium-lithium diode detector in the background.

Figure 9. A profile of Cs-137 concentrations measured in a sediment core. The concentration maximum indicates the year 1963 and the depth at which Cs-137 disappears marks the year 1954.
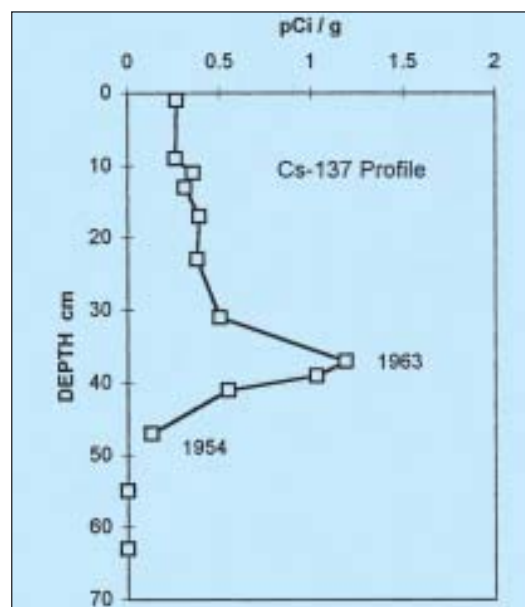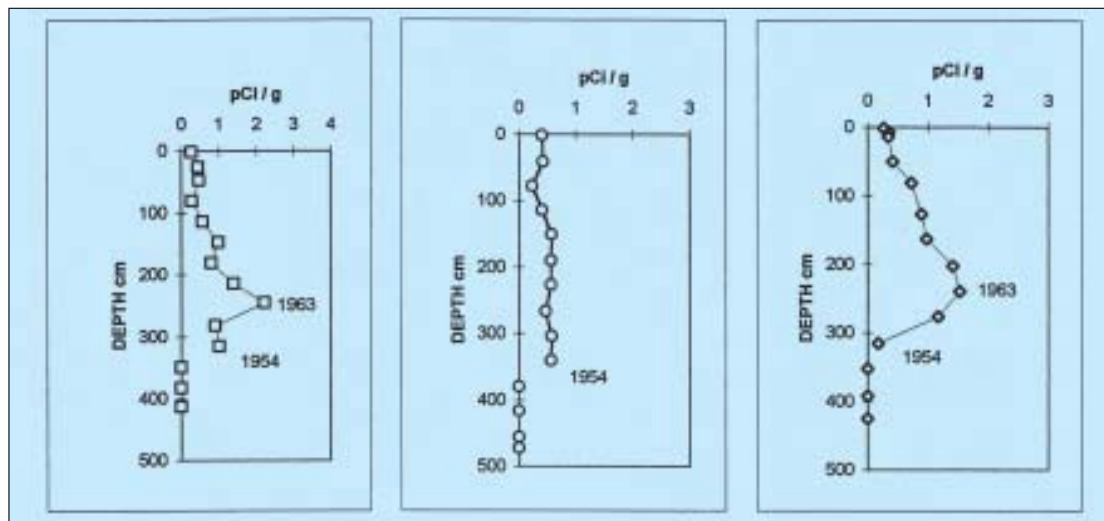
Figure 10. Profiles of Cs-137 in sediments exhibit different shapes. Overall concentration levels and mixing of the surface sediments are contributing factors.

It is often useful to compare sedimentation rates calculated by the Pb-210 method and the Cs-137 method. The core illustrated in Figure 9 is the same core which was illustrated for Pb-210 in Figure 1. Analysis of the Pb-210 profile resulted in a calculated sedimentation rate of 0.95 cm/y, which is lower than the rate of 1.2 cm/y calculated from both markers of the Cs-137 profile. The discrepancy is explained by the different time frames characterised by the two methods. The sedimentation since 1954 (characterised by the Cs-137 method) has been more rapid than the 100 year average characterised by the Pb-210 method.

The shapes of Cs-137 profiles vary significantly (Figure 10). Cases commonly occur where a Cs-137 horizon is found marking the year 1954, but no maximum concentration is found to mark the year 1963. This often happens at locations where all the Cs-137 concentrations are low or near the detection limit. Other cases show more than one maximum in concentration, although the larger maximum generally marks the year 1963. The shape of the Cs-137 maximum can be sharp and distinct or broad and blunt. This shape has been related to the depth of surface mixing of sediments by biological organisms or by physical processes. Modelling studies show that broader maxima in Cs-137 concentrations are produced when several cm of surface sediment are mixed (Ref. 4).

CHRONOLOGY STUDIES OVER THE LAST HALF YEAR: THE BE-7 METHOD

Several radioactive species are continually produced in the atmosphere by the interaction of cosmic radiation (from outer space) with gas molecules. One of these species is Be-7 (beryllium-7) which was confirmed by measurements of rain water (Ref. 10). This nuclide forms part of the natural fallout which accumulates in the surface sediments of water bodies. It is used to provide additional information in surveys for chronology purposes.

It is practical to measure Be-7 by direct gamma spectral analysis of a sediment sample simultaneously with the Cs-137 measurement, without additional cost. These two radioactive species produce gamma radiation at different energies, so that they appear in different regions of the spectrum (Figure 8). Such measurements are not as sensitive, however, as the more laborious chemical separation procedures which are sometimes used in scientific studies.

Table II. Cs-137 concentrations plotted in Figure 9.

Depth cm	Cs-137 pCi/g
0 – 2	0.27 +– 0.11
8 – 10	0.26 +– 0.08
10 – 12	0.36 +– 0.11
12 – 14	0.31 +– 0.12
16 – 18	0.39 +– 0.11
22 – 24	0.38 +– 0.10
30 – 32	0.51 +– 0.10
36 – 38	1.19 +– 0.11
38 – 40	1.03 +– 0.11
40 – 42	0.55 +– 0.11
46 – 48	0.13 +– 0.07
54 – 56	< 0.09
62 – 64	< 0.09

Tolerances of the measurements are based on detection uncertainties at the 2 sigma (95% confidence) level.

Beryllium-7 decays relatively quickly, with a half-life of 53 days (in one half-life the concentration of a radioactive species decays to half of its original level). For comparison, the half-life of Pb-210 is 22 years and that of Cs-137 is 30 years. Because of its rapid decay, Be-7 is only found in the first few cm of sediments. Consequently, it has limited utility in the practical study of sedimentation rates for characterising water systems. It has been used effectively, however, for the scientific study of vertical mixing processes in surface sediments (Ref. 11).

For practical survey work, the primary utility of Be-7 is to indicate whether there has been very recent deposition at a sampling location (within the last half year) and whether the sediment core has been taken with the surface sediments intact. Detection of Be-7 in the first cm or two of a core gives affirmative answers to these questions. But conversely, if Be-7 is not detected in the surface sediments, these questions are not necessarily answered in the negative. The reason for this is that vertical mixing of the surface sediments by biological organisms or by physical processes can dilute the Be-7 concentration to the point where it is undetectable by the practical, direct gamma analysis method.

To illustrate, in two different studies of the Passaic River system, one study showed reliable detection of Be-7 in 38% of the cores taken. In the other study, only 16% of the cores showed Be-7 near the surface. It is evident that Be-7 can provide supplemental information in a practical survey, but that it is not as useful as Pb-210 or Cs-137 for measuring sedimentation rates (Ref. 8).

Conclusion

In recent years, many surveys of contaminated river systems have included analyses of sediment cores to provide chronology information. These analyses have been used to calculate sedimentation rates and to provide calendar dates associated with various levels in the sediments. Chronology information of this kind can assist in determining which industries caused the contamination, for industrial production, or release records can be compared to the dates of buried materials. Chronology information is also useful for determining whether buried substances are migrating or degenerating, knowledge which is often useful in planning dredging operations.
The techniques used in sediment chronology are derived from universities and from scientific studies. Although many techniques have been developed, only measurements of man-made Cs-137 and natural Pb-210 and Be-7 have emerged as practical approaches to the chronology of sediments accumulating within the last 100 years for site characterisation and remediation studies.

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